Multiple nonlinear Bragg diffraction of femtosecond laser pulses in $\chi^{(2)}$ photonic lattice with hexagonal domains

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Abstract. Frequency doubling of femtosecond laser pulses in a two-dimensional rectangular nonlinear photonic lattice with hexagonal domains is studied experimentally and theoretically. The broad fundamental spectrum enables frequency conversion under nonlinear Bragg diffraction for a series of transverse orders at a fixed longitudinal quasi-phase-matching order. The consistent nonstationary theory of frequency doubling of femtosecond laser pulses is developed using the representation based on the reciprocal lattice of the structure. The calculated spatial distribution of the second-harmonic spectral intensity agrees well with the experimental data. Condition for multiple nonlinear Bragg diffraction in 2D nonlinear photonic lattice is offered. The hexagonal shape of domains contributes to multibeam second harmonic excitation. The maximum conversion efficiency for a series of transverse orders in the range from 0.01% to 0.03% is obtained.

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1. Introduction

For the last decade, the nonlinear Raman-Nath diffraction (NRND) has attracted much attention [1, 2, 3, 4, 5, 6, 7]. This phenomenon represents a series of second harmonic beams resulting from multiples of the primary reciprocal lattice vector implemented by the periodical modulation of the sign of second-order nonlinear susceptibility of a structure in the transverse direction. Usually, the longitudinal phase-matching conditions for the NRND orders are unmet and the second-harmonic (SH) intensity oscillates along the propagation direction, which results in the low conversion efficiency [4, 5]. As was reported in [3], the second harmonic generation (SHG) efficiency can be significantly increased by varying the angle of incidence of the fundamental frequency (FF) beam. In this case, the Cerenkov nonlinear diffraction described by the longitudinal phase matching can enhance the corresponding NRND order, which results in the strong SHG under the nonlinear Bragg diffraction (NBD). At the same time, it seems promising to increase the NRND by the quasi-phase-matching with the use of two-dimensional (2D) nonlinear photonic lattices. They are found to be promising for multiwavelength generation [8], implementation of broadband parametric interactions [9], producing path-entangled photons [10] and multiple copies of beams carrying orbital angular momentum [11], observation of nonlinear Talbot effect [12, 13]. Up to now, the 2D nonlinear photonic lattices with the periodic [14], randomaized [15], superimposed [16], and chirped [17] modulation of the nonlinear susceptibility sign in the propagation direction have been investigated. Among them, only periodic structures provide a specific reciprocal lattice vector for all the NRND orders, while the phase mismatches for different orders are different. As a consequence, the respective phase mismatches are compensated at different wavelengths. This can be made by using an appropriate bandwidth of the fundamental spectrum, which is ensured by femtosecond laser pulses. In this case, the nonlinear Bragg diffraction can take place for specific spectral components within the spectral width provided by a laser. Previously, we studied the second harmonic generation of femtosecond laser pulses under the NRND and Cerenkov nonlinear diffraction in a 1D nonlinear photonic lattice [5, 18]. In [19, 20, 21, 22], the quasi-phase-matched SHG in the continuous wave regime in the 2D nonlinear photonic lattices was investigated. The frequency doubling of femtosecond laser pulses in the 2D nonlinear photonic lattice of a special design was investigated by us in [14]; however, this problem remains unsolved for the 2D nonlinear photonic lattices with the translational symmetry and arbitrary domain shape.

In this Letter, we report the results of experimental and theoretical study of the multiple SHG of femtosecond laser pulses in the 2D nonlinear photonic lattices representing rectangular lattice with hexagonal domains. The spectral width of femtosecond pulses allows the process to be implemented under quasi-phase-matching, which results in the multiple nonlinear Bragg diffraction at specific wavelengths. The choice of the hexagonal shape of domains are caused by the intrinsic property of the lithium niobate, which is that the domains have a tendency to take the form of hexagonal prisms. Moreover, the hexagonal shape of domains provides nearly equal intensities for several second harmonic peaks, as it will be shown further.

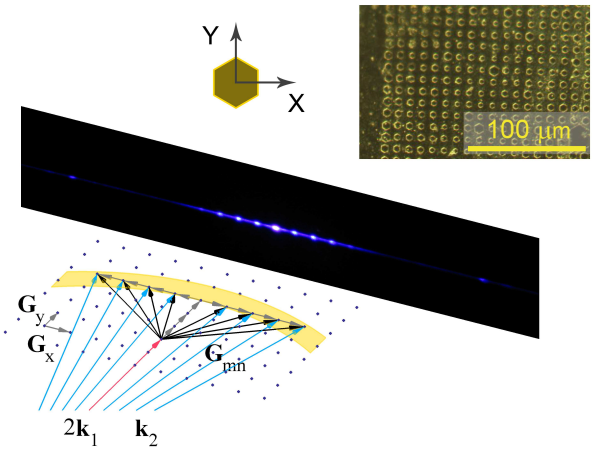

Figure 1. Far-field image of the experimental SH pattern and vectorial phase-matching diagram in the reciprocal space. The reciprocal space region corresponding to the laser spectral bandwidth is colored with yellow. Inset: Optical image of the etched CLN surface.

2. Theoretical model

Two-dimensional nonlinear photonic lattice provides discrete set of reciprocal lattice vectors resulting in quasi-phase-matched SHG at several directions as

shown in figure 1. The spatial distribution of nonlinear susceptibility within a 2D nonlinear photonic lattice can be expressed as the series [19]

$$g(x,y)=\sum_{m,n=0,\pm1,\ldots} g_{mn}\mathrm{e}^{-\mathrm{i}(mG_x x+nG_y y)}, \tag{1}$$

where g_{mn} are Fourier coefficients, $G_{x,y}=2\pi/\Lambda_{x,y}$ are the primitive vectors in the reciprocal space (figure 1), i.e., the primitive reciprocal lattice vectors ($\Lambda_{x,y}$ is the nonlinearity modulation period along the corresponding coordinate) and m and n are the longitudinal (QPM) and transverse (NRND) orders ($m,n\in\mathbf{Z}$), respectively. For the rectangular lattice with the hexagonal domains, the Fourier coefficients are (compare with [19, 20])

$$\begin{aligned} g_{mn}&=\operatorname{sinc}\left(m\frac{G_x\Lambda_x}{2}\right)\operatorname{sinc}\left(n\frac{G_y\Lambda_y}{2}\right)+\frac{2\sqrt{3}}{nG_y}\frac{d}{\Lambda_x\Lambda_y}\\ &\times\left\{\operatorname{sinc}\left[\frac{d}{4}(\sqrt{3}mG_x+nG_y)\right]\sin\left[\frac{d}{4}(\sqrt{3}mG_x-3nG_y)\right]\right.\\ &\left.-\operatorname{sinc}\left[\frac{d}{4}(\sqrt{3}mG_x-nG_y)\right]\sin\left[\frac{d}{4}(\sqrt{3}mG_x+3nG_y)\right]\right\}, \end{aligned} \tag{2}$$

where d is the hexagonal edge length, $\operatorname{sinc}(x)=\sin(x)/x$.

In the quasi-optical approach, the SH field evolution inside the structure under the slowly varying amplitude approximation in the low-conversion regime is governed by the equation [23]

$$\begin{aligned} &\left(\frac{\partial}{\partial y}+\frac{1}{u_2}\frac{\partial}{\partial t}\right)A(t,r,y)+\frac{\mathrm{i}}{2k_2}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)A(t,r,y)\\ &=\Gamma g(x,y)F_r^2(r)F_t^2\left(t-\frac{y}{u_1}\right)\mathrm{e}^{\mathrm{i}\Delta k y}, \end{aligned} \tag{3}$$

where $\Delta k=k_2-2k_1$, $\Gamma=-\mathrm{i}\beta_2 I_1$, $\beta_2=2\pi k_2\chi^{(2)}/n_2^2$, I_1 is the FF intensity at the beam center, and u_2 is the SH group velocity. For the Gaussian distribution of intensity in the time and transverse coordinates, the functions $F_t^2(t)$ and $F_r^2(r)$ take the forms

$$\begin{cases} F_r^2(r)=\exp\left(-2r^2/a^2\right)\\ F_t^2(t)=\exp\left(-2(t-y/u_1)^2/\tau^2\right). \end{cases}$$

Here, u_1 is the FF group velocity and 2τ and $2a$ are the pulse duration and the focal spot diameter of the FF beam, respectively.

The solution of (3) can be presented in the form

$$\begin{aligned} &A(\Omega,K_x,K_y,K_z,y)=\\ &\alpha y\sum_{mn}g_{mn}\exp\left(-\frac{a^2}{8}\left[(K_x+mG_x)^2+K_z^2+\frac{\Omega^2\tau^2}{a^2}\right]\right)\\ &\times\operatorname{sinc}\left(\frac{y}{2}\left[\Delta k+\nu\Omega-\frac{K_x^2}{2k_2}+nG_y\right]\right)\\ &\times\exp\left(\frac{\mathrm{i}y}{2}\left[\Delta k-\left(\frac{1}{u_2}+\frac{1}{u_1}\right)\Omega+\frac{K_x^2}{2k_2}+nG_y\right]\right), \end{aligned} \tag{4}$$

where $\alpha=\Gamma\tau a^2(\pi/2)^{3/2}$, $\nu=1/u_2-1/u_1$ is the group velocity mismatch, $\Omega=2\pi c(1/\lambda_2-2/\lambda_0)$ is the frequency detuning from the central double frequency, λ_0 is the central FF wavelength, λ_2 is the SH spectral component, and K_x and K_z are spatial frequencies. It is worth noting that in the stationary regime (4) reads to the form [21]. Additionally, derived expression (4) can be implemented for the calculation of SHG in 2D rectangle nonlinear photonic lattices with arbitrary domain shape. The requirement is to specify the Fourier coefficients g_{mn}.

Consider the spectral density for fixed values of m and n at the distance $y=L$

$$\begin{aligned} &S_{mn}(\Omega,K_x,K_y,y=L)=|A_{mn}(\Omega,L)|^2=\\ &\alpha g_{mn}L\exp\left[-\frac{(\tau\Omega)^2}{4}-\frac{a^2}{4}\left(K_x+mG_x\right)^2\right]\\ &\exp\left[-0.09\left(\Delta k+\nu\Omega-\frac{K_x^2}{2k_2}+nG_y\right)L\right]. \end{aligned} \tag{5}$$

Here the substitution $\operatorname{sinc}^2(\xi)\to\exp(0.36\xi^2)$ is used [23] and $K_z=0$. The first exponent in (5) describes NRND and condition $K_x+mG_x=0$ defines its orders. When the QPM is introduced in the longitudinal direction, i.e. the condition

$$\Delta k+\nu\Omega-\frac{K_x^2}{2k_2}+nG_y=0 \tag{6}$$

is fulfilled the nonlinear Bragg diffraction occurs.

Analysis of (5) shows that the spectral position of the nonlinear Bragg diffraction order $\{m,n\}$ can be determined as

$$\Omega_{mn}=\nu^{-1}\left(\frac{Q}{2L}m^2-\frac{2\pi}{\Lambda_y}n-\Delta k\right). \tag{7}$$

Here $Q=2\pi\lambda_2 L/n_2\Lambda_x^2$. It can be seen that Ω_{mn} obeys the quadratic law on the transverse order m. The first term in (6) increases slower than the second one at not very large values Q as $L\gg\Lambda_y$ or, since $\lambda_2/\Lambda_x\ll1$, in the largest of all the experimental implementations. It means also that the spectral intervals between the adjacent orders m are smaller than spectral intervals between the orders n. Using (6), we can find the required lattice period in the longitudinal direction in order to obtain the multiple quasi-phase-matched SHG at a given wavelength.

The spectral interval between the peak orders $m+1$ and m is

$$\Delta\lambda_m=\frac{(1+2|m|)\lambda_2^3}{2n_2c|\nu|\Lambda_x^2}=\frac{(1+2|m|)\lambda_2^2 Q}{4\pi c|\nu|L}. \tag{8}$$

The multiple nonlinear Bragg diffraction orders are observed when $\Delta\lambda/\Delta\lambda_m\gg1$ ($\Delta\lambda$ is the fundamental frequency spectral width).

The spectral interval between the adjacent orders $n+1$ and n is

$$\Delta\lambda_n=\frac{\lambda_2^2}{c|\nu|\Lambda_y}. \tag{9}$$

Note that $\Delta\lambda_n$ is independent of the order n and amounts approximately to 40 nm under the experimental conditions. The spectral width of an arbitrary order $\{m,n\}$ can be found as

$$\delta\lambda_{mn} \approx 2\sqrt{\ln 2}\frac{\lambda_2^2}{c|\nu|L}. \quad (10)$$

Let us define the SH power in the NBD order $\{m,n\}$ at the frequency Ω_{mn} so that $P_{mn} = P(\Omega_{mn};L)$, which is governed by

$$P_{mn} \approx \int_{-\infty}^{\infty} S_{mn}(\Omega=\Omega_{mn}+\delta\Omega;L)d\delta\Omega$$
$$= \frac{2\sqrt{\pi}(\alpha g_{mn}L)^2}{\tau\sqrt{1+(\tau_d/\tau)^2}}\exp\left[-\frac{(\tau_d/\tau)^2}{4(1+(\tau_d/\tau)^2)}(\tau\Omega_{mn})^2\right], \quad (11)$$

where $\tau_d = 0.6|\nu|L$ is the group delay time. Then, the ratio between SH powers $\eta = P_{m_1n_1}/P_{m_2n_2}$ for different pairs $\{m,n\}$ under the condition $\tau_d \gg \tau$ is

$$\eta = \left(\frac{g_{m_1n_1}}{g_{m_2n_2}}\right)^2 \frac{2\sqrt{\pi}}{\tau_d}\exp\left[-\frac{1}{4}\tau_d^2(\Omega_{m_1n_1}^2-\Omega_{m_2n_2}^2)\right]. \quad (12)$$

If the factor of the exponent in (12) is smaller than unity, the ratio between SH powers is determined as the squared ratio between the 2D Fourier coefficients of nonlinear photonic lattice.

3. Results and discussion

In the experiment, we used a congruent lithium niobate-based nonlinear photonic lattice. The nonlinear photonic lattice structure was fabricated by electric field poling (Labfer Ltd) [24]. The sample was $5.0 \times 2.0 \times 0.5$ mm^3 ($x \times y \times z$) in size. The square lattice period was about $\Lambda_{x,y} = 10$ μm. Each lattice point contains domains with the nearly hexagonal shape (see the inset in figure 1). One of the hexagonal domain edges was parallel to the y axis of the crystal. The measured hexagonal edge length d was measured to be $\sim 0.29\Lambda_x$. In the experiments, the fundamental frequency beam from a Spectra-Physics Tsunami Ti:sapphire oscillator delivering 85-fs pulses at a repetition rate of 80 MHz was focused onto the sample by a 20-cm lens to produce a beam with a focal diameter of $2a = 136$ μm (confocal parameter 3.4 cm). The fundamental radiation propagated along the y axis and its polarization coincided with the z axis to employ the highest nonlinear coefficient of lithium niobate d_{33}. The central fundamental wavelength has been scanned from 830 to 880 nm to obtain multiple SHG under NBD. Figure 2 shows the SH far-field images observed on a screen distant by 7 cm from the sample for a set of the central fundamental wavelengths. NBD is clearly observed at the wavelengths 850-860 nm (figure 2(b-d)). At the central fundamental wavelength 856 nm is chosen as an optimal wavelength since the maximum SHG efficiency is achieved for a larger number of transverse orders m in this case (figure 2(c)). Therefore, in the following experiments the central fundamental wavelength was tuned to a wavelength of 856 nm to implement the SHG under the 3^{rd} order quasi-phase-matching for the 2^{nd} transverse order (the NBD order $\{2,3\}$).

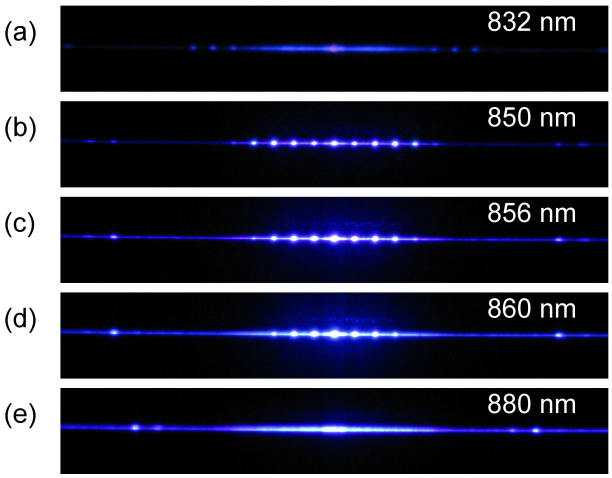

Figure 2. Far-field image of the experimental SH pattern on central FF wavelength.

The second harmonic spectra were measured by an MSDD 1000 spectrometer (TII, Solar) with a spectral resolution of 0.02 nm. It can be seen in figure 2 that the SH spectra consist of a single peak that undergoes the short-wavelength spectral shift with increasing order. Moreover, the central peak is surrounded by secondary maxima for $m = 0$ and $m = \pm 1$. According to the Fourier analysis of the structure (see the inset in figure 2), the secondary maxima originate from periodic modulation of nonlinearity in the structure. The hexagonal shape of domains is analyzed to provide distribution of intensity between NBD orders. In addition, some background appears in the spectral intensity, which can be attributed to the random variations in the domain size (figure 3).

The angular dependence of the SH spectral intensity ($S_2(\Omega,K) = |A(\Omega,K,L)|^2$) calculated using (4) is shown in Fig. 4. Good agreement between calculated dependence and measured SH spectra is achieved under the following parameters: the sample length $L = 2$ mm, the hexagonal edge length $d = 2.9$ μm, the central FF wavelength 856.4 nm, the pulse duration $2\tau = 144$ fs, the focal spot diameter $2a = 136$ μm. The required refractive index data were taken from [25]. The approach proposed allows us not only to describe the angular distribution of the spectral intensity, but also to evaluate the spectral and angular widths of SH peaks for specific NBD orders. Additionally, the calculated dependence demonstrates the short-wavelength spectral shift of maxima with

increasing NBD order m according to (7). Unlike the calculated angular dependence of spectral intensity, the measured SH spectra are broaden, which can be attributed to the fluctuation of hexagonal domain sizes. The angular distribution of the SH intensity integrated over the spectrum is shown in the same figure in comparison with the measured dependence obtained by translating the narrow width slit in the transverse direction. For this purpose, a Newport 918D power meter was also used.

The SH peak wavelengths (λ_m), spectral widths ($\delta\lambda_m$), and intervals ($\Delta\lambda_m$) were calculated using (8), (9) and (10), respectively. These values for $m \in [-4, 4]$ are given in table 1. In addition, the Table 1 summarizes the average measured SH peak wavelengths, spectral widths, and intervals. The measurement accuracy of spectral characteristics was about 0.02 nm. It can be seen that the calculated values are in good agreement with the measured ones.

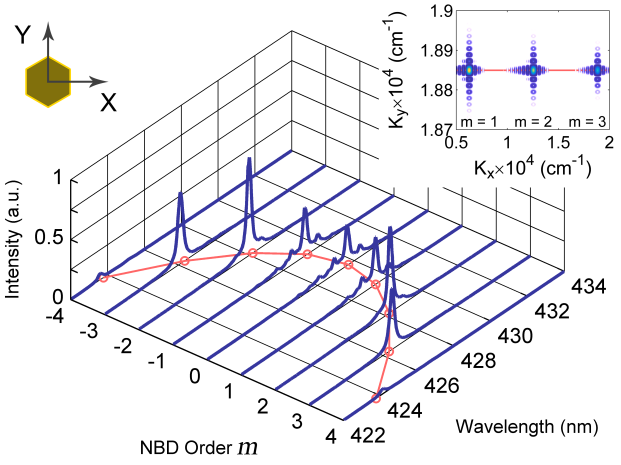

Figure 3. Measured SH spectra and SH peak wavelengths (red points) for the NBD orders. Inset: Fourier spectrum of the structure under study (the red solid line indicates the value $K_y = 3G_y$).

As can be seen in figure 1, the central spots corresponding to the RNND are brighter than the side spots corresponding to the Cerenkov nonlinear diffraction [14]. The measured maximum conversion efficiency for a series of transverse orders (central spots) lies in the range of $0.01-0.03\%$, which is higher than the value obtained in our previous work [14] by about twenty times of magnitude. Let us introduce the structure gain coefficient as a ratio of employed longitudinal orders n and structure lengths L, i.e. $(n_1 L_2/n_2 L_1)^2$, where the indexes $1, 2$ are related to the previous [14] and the current study, respectively. For $m = \pm 1$, calculation gives the structure gain coefficient ~ 18, which agrees with our measurements.

Usually, acoustooptics distinguishes the two diffraction regimes: Raman-Nath and Bragg. The

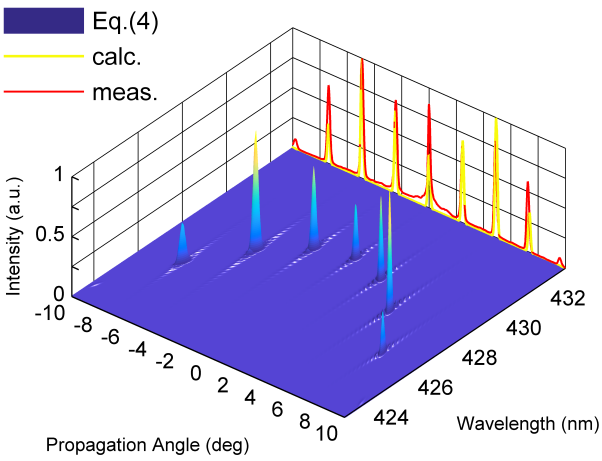

Figure 4. Angular dependence of the SH spectral intensity calculated using (4) and calculated and measured angular dependences of the SH intensity.

criterion of the diffraction regime is the Klein-Cook parameter Q_{KC} [26]. At $Q_{KC} \gg 1$, the Bragg diffraction takes place; otherwise, the Raman-Nath diffraction is observed ($Q_{KC} < 1$). Assuming the diffraction phenomena to have the same nature, we have introduced the similar parameter to distinguish the nonlinear diffraction regime. In our model, it is the parameter Q in (6). Calculations yield a value of $Q \approx 24$, which indicates the Bragg nonlinear diffraction regime.

4. Conclusion

Thus, frequency doubling of femtosecond laser pulses in a 2D rectangular nonlinear photonic lattice with hexagonal domains was studied. It was shown that the broad fundamental spectrum enables frequency conversion under the nonlinear Bragg diffraction for five transverse orders with the efficiency varying from 0.01% to 0.03%, which is higher than the value obtained in our previous work by about twenty times of magnitude. The consistent nonstationary theory of frequency doubling of femtosecond laser pulses was developed, which uses the representation based on the reciprocal lattice of the structure. Condition for nonlinear Bragg diffraction in 2D nonlinear photonic lattice is suggested. The calculated spatial distribution of the second harmonic spectral intensity agrees well with the experimental data.

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Table 1. Calculated ((10) and (8)) and experimental spectral widths ($\delta\lambda_m$) and intervals ($\Delta\lambda_m$). The brackets $\langle ...\rangle$ indicate the average value.

m	λ_m (nm)	$\delta\lambda_m$ (nm)	$\Delta\lambda_m$ (nm)	$\langle\lambda_m^{exp}\rangle$ (nm)	$\langle\delta\lambda_m^{exp}\rangle$ (nm)	$\langle\Delta\lambda_m^{exp}\rangle$ (nm)
0	429.7	0.33	-	429.6	0.2	-
$\pm$ 1	429.3	0.33	0.37	429.3	0.2	0.3
$\pm$ 2	428.2	0.33	1.12	428.1	0.3	1.2
$\pm$ 3	426.3	0.33	1.85	426.3	0.3	1.8
$\pm$ 4	423.8	0.33	2.56	423.8	0.4	2.5

Government of the Russian Federation (Act 211, Agreement 02.A03.21.0006) for fabrication of 2D nonlinear photonic lattice in lithium niobate crystals.

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